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# 150-ps broadband low dispersion mirror thin film damage competition

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## ABSTRACT

Broadband low dispersion mirrors are fluence-limiting and pulse-shape-limiting components in short pulse lasers. To better understand the current technology state of broadband low dispersion mirrors, a laser damage competition was held at the 2015 Laser Damage Conference. Participants were asked to submit mirrors that met a minimum reflection of 99.5% at 45 degrees incidence angle at “P” polarization with a Group Dispersion Delay (GDD) of  $<\pm 100 \text{ fs}^2$  over a spectral range of  $773 \text{ nm} \pm 50 \text{ nm}$ . The participants selected the coating materials, design, and deposition method. Laser damage testing was performed using the raster scan method with a 150 ps pulse length on a single testing facility to enable a direct comparison among the participants. GDD measurements were performed to validate specification compliance. Unfortunately nearly half of the submitted samples were found to not meet the GDD specifications. Details of the deposition processes, cleaning method, coating materials, and layer count are also shared.

**Keywords:** laser damage, laser damage testing, thin film, broadband low dispersion mirror, multilayer, picosecond pulse length

## 1. INTRODUCTION

There are currently over 50 petawatt class lasers worldwide.<sup>1</sup> These laser systems are used for a number of research projects ranging from inertial confinement fusion, radiography, particle acceleration, studying materials at high temperatures and pressures, radiation therapy, secondary source generation, and medical isotope creation to name a few. There is a huge growth in this field considering that in 1998 the only petawatt class laser in the world was on the NOVA laser at Lawrence Livermore National Laboratory.<sup>2</sup> Pulse compression gratings and short pulse transport mirrors remain one of the fluence limiting components on these laser systems.

## 2. PARTICIPATION

Thirty-three samples were submitted by sixteen different participants representing six different countries as observed in table 1 below. The samples were manufactured by each participant on their own 50 mm diameter by 10 mm thick substrates and submitted for laser damage testing and GDD measurements. Five participants were new to this series of thin film laser damage competitions that started in 2008. Laser Zentrum Hannover remains the only institute that has participated in every competition. In addition to providing the samples, the participants were required to supply the following information:

- Number of coating layers
- Coating materials
- Reflectance scans over the specified spectral bandwidth
- A brief description of the deposition method
- A brief description of the cleaning method
- Substrate material

Table 1 List of sixteen participants, counties of origin, and number of years participating in the Laser Damage Conference thin film damage competition.

<b>Company / Institute</b>	<b>Country</b>	<b>Years of participation</b>
Advanced Thin Films	USA	3
Aerospace Times Laser Inertial Technology	China	1
Arrow Thin Films	USA	2
Carl Zeiss	Germany	1
Colorado State University	USA	1
CVI Melles Griot	USA	3
Laser Components	Germany	6
Laser Zentrum Hannover, e.V.	Germany	8
Nikon	Japan	5
Optical Coatings Japan	Japan	2
Optida UAB	Lithuania	2
Research Electro-Optics	USA	1
Sandia National Laboratory	USA	3
Schott	USA	2
SLS Optics	Great Britain	4
Tongji University	China	4

### 3. SAMPLES

Samples were assigned a unique two-digit participant code to maintain sample anonymity. The first digit consisted of a letter ranging from A to Q for the sixteen participants respectively. The second digit was a sample number ranging from 1 to 4 depending on how many samples were supplied by each participant. The connection between the participant name and code was unknown to the damage testing service and GDD measurement service. They only had access to the participant code so as to remain unbiased and to protect the identities of participants whose samples had lower laser resistance or non-compliant GDD. Only the participant code is used in this paper and also the talk at the Laser Damage conference to maintain participant anonymity.

The coatings had to meet the following specifications:

- Wavelength range  $773 \pm 50$  nm
- Incidence angle 45 degrees
- Polarization “P”
- Reflectance  $> 99.5\%$
- Group Dispersion Delay (GDD)  $< \pm 100$  fs<sup>2</sup>
- Ambient environmental conditions
  - Temperature ( $20 \pm 2$  degrees C)
  - Relative humidity ( $40 \pm 20\%$ )
- No reflected wavefront or stress requirement
- No surface quality requirement

Five of the last seven laser damage competitions involved multilayer coatings that were tested in the near infrared with coatings that were high reflectors<sup>3-4</sup>, polarizers<sup>5</sup>, and Fabry-Perot filters<sup>6</sup>. One of the mirrors was tested at 150 fs while

the other coatings were tested at nanosecond pulse lengths. Over these five thin film damage competitions, every winning entry was composed of hafnia and silica. Four of these five samples were laser damage tested with nanosecond scale pulses and all of the winning coatings were manufactured by e-beam (one included ion assist). Therefore, the specifications for this competition were intentionally selected to minimize the chance of a simple quarter-wave coating design of hafnia and silica deposited by e-beam to meet the spectral and GDD requirements over the entire spectral bandwidth. Figure 1 illustrates the spectral and GDD bandwidth as a function of refractive index for the high refractive index material of a 50-layer quarter wave stack design with silica as the low index material. These calculations were done with Essential Macleod,<sup>7</sup> a thin film design program, using the refractive indices provided within the software. The refractive index of coating materials is deposition process dependent, so the values in the chart are representative of typical e-beam deposited films.

Because of the spectral and GDD bandwidth requirements, a significant fraction of the coatings utilized a design strategy that had two different high index coating materials. Typically alternating layers of the highest refractive index material and silica layers are deposited on the substrate first to achieve the spectral bandwidth. Alternating layers of higher laser resistant hafnia and silica layers are then deposited on the top of the coating to reduce the electric field going into the lower laser resistant higher refractive index materials. Table 2 lists the material selections used for this competition and table 3 lists the deposition processes. Of particular note is that two thirds of the submitted samples all contain hafnia as a high index coating material. Two of the samples that were submitted were deposited by co-evaporation of hafnia and silica resulting in a mixed high index material.

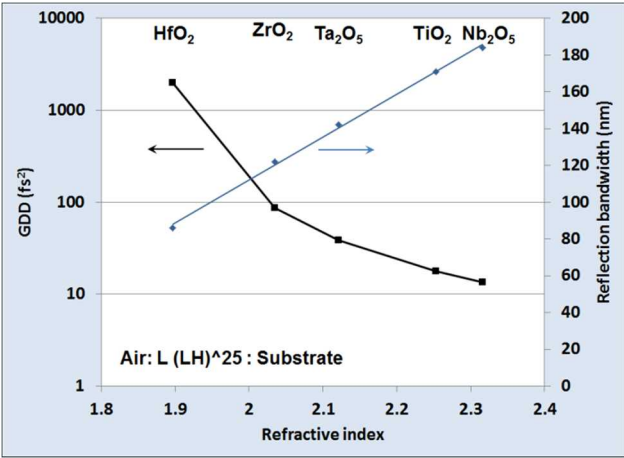


Fig. 1 Reflection and GDD bandwidth as a function of high refractive index material.

Table 2 Sample distribution as a function of high refractive index coating materials.

High refractive index materials	Number of samples
Ag, Cu, & HfO <sub>2</sub>	1
Ag & HfO <sub>2</sub>	1
HfO <sub>2</sub>	8
HfO <sub>2</sub> and SiO <sub>2</sub> (mixture)	2
HfO <sub>2</sub> & Nb <sub>2</sub> O <sub>5</sub>	2
HfO <sub>2</sub> & Ta <sub>2</sub> O <sub>5</sub>	4
HfO <sub>2</sub> & TiO <sub>2</sub>	4
Ta <sub>2</sub> O <sub>5</sub>	6
TiO <sub>2</sub>	2
ZrO <sub>2</sub>	2
Unknown	1

Table 3 Sample distribution as a function of coating deposition process.

Deposition process	Number of samples
Electron-beam (e-beam)	8
Ion Assisted Deposition (IAD)	8
Ion Beam Sputtering (IBS)	13
Magnetron Sputtering	3
Unknown	1

#### 4. MEASUREMENTS

The samples were laser-damage tested at the Femtosecond Solid Dynamics Lab at the Ohio State University using the raster scan method described by Kafka<sup>8</sup> and Borden<sup>9</sup>. Damage testing was performed at 780 nm with a centered spectral bandwidth of 35 nm. The repetition rate was 500 Hz with TEM<sub>00</sub> pulses. The samples were raster scanned over a 3 mm by 3 mm area starting at a fluence of 1 J/cm² and increasing in 2 J/cm² increments. A new area was scanned at each higher fluence to minimize the potential for laser conditioning. The beam diameter was 82 microns at 1/e². With a

scanning speed of 0.625 mm/s and repetition rate of 500 Hz, each site was exposed to roughly 15 shots at 90% peak fluence allowing the opportunity to grow the laser damage for easier detection. An in-situ camera was used for damage detection. A diagram of the damage test set up is shown in figure 3. The accuracy of the fluence was 5 percent.

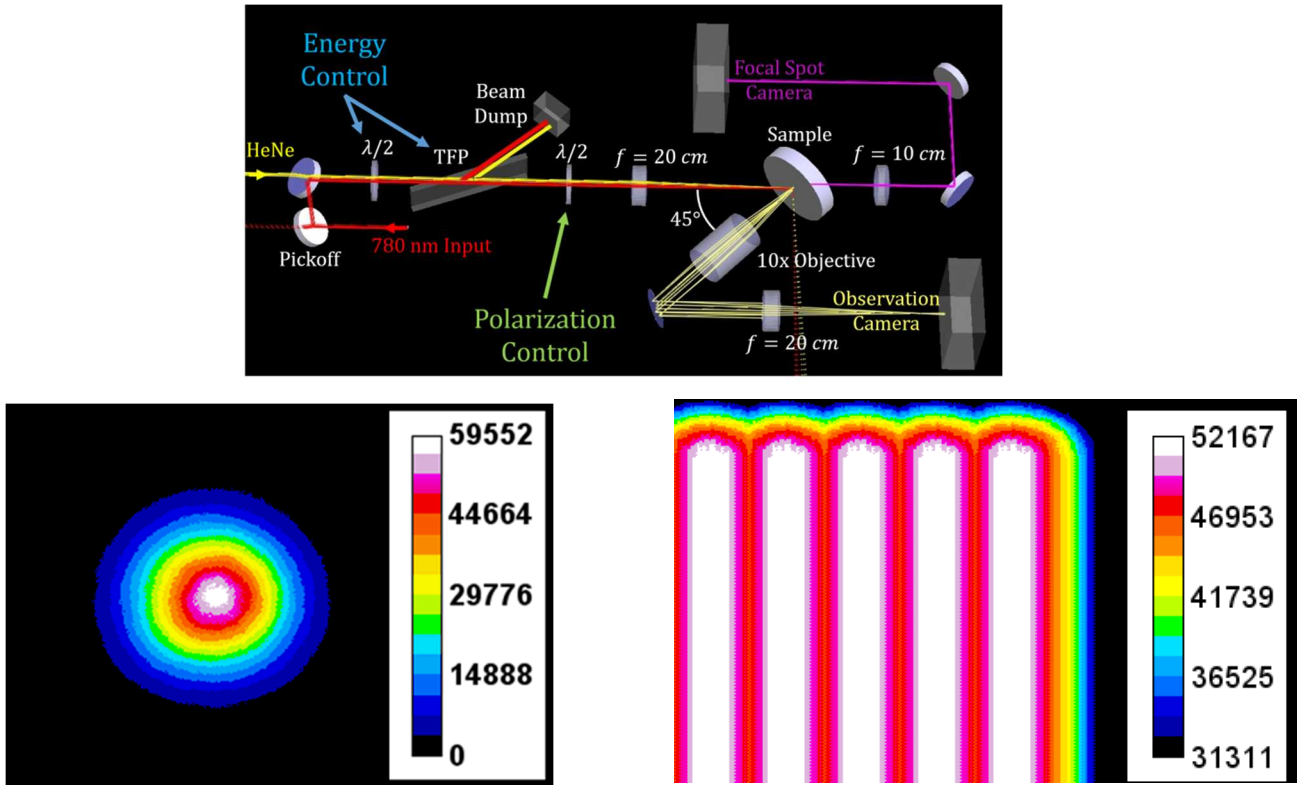


Fig. 3 Schematic of the laser damage test setup (top image), beam profile (bottom left image) and raster scan pattern (bottom right image).

GDD measurements were performed at KMLabs in Boulder, Colorado on the Chromatis instrument depicted in figure 4. Measurements were done over a wavelength range of 700 – 850 nm. A gold calibration standard was used to establish a measurement error of less than 5 fs<sup>2</sup>. The beam diameter was 6 mm. GDD measurements were performed after damage testing to minimize possible handling- and contamination-induced laser resistance degradation of the samples.

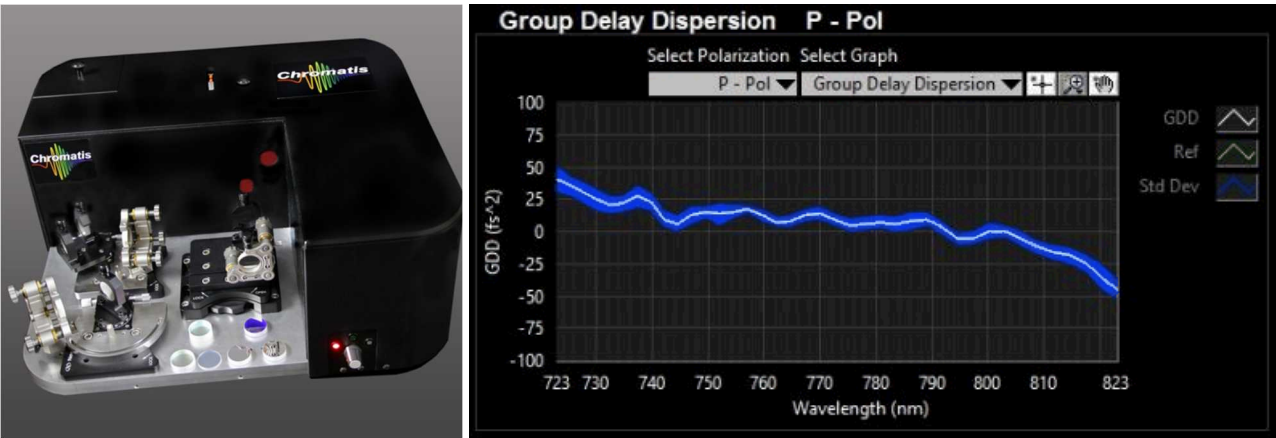


Fig. 4 Chromatis instrument used for measuring GDD (left image) & GDD output of the winning entry (right image).

## 5. RESULTS

The results of the GDD measurements are plotted in figure 5. Unfortunately only nineteen of the samples fully met the specification of  $\pm 100 \text{ fs}^2$  over a wavelength range of  $773 \pm 50 \text{ nm}$ . Of the fourteen samples that failed the GDD specification, three of them had insufficient bandwidth to cover a full 100 nm of spectral bandwidth regardless of spectral centering. With proper centering, the remaining eleven samples would have met the bandwidth requirement. These results imply that GDD modeling alone is insufficient to validate compliance with specifications. GDD measurements must also be performed.

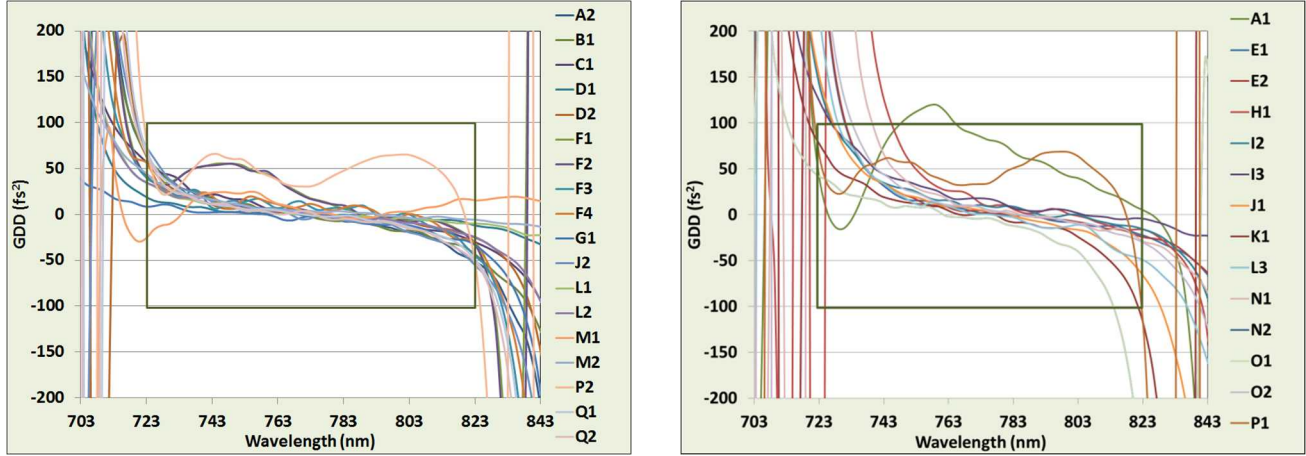


Fig. 5 GDD results of the samples that pass specification within the red rectangular box (left image) and samples that failed the specifications (right image).

As expected, there was no strong correlation between either GDD or layer count on the laser resistance of the coating as illustrated in figure 6. Like previous thin film laser damage competitions, a much stronger correlation between coating materials and deposition processes was observed.

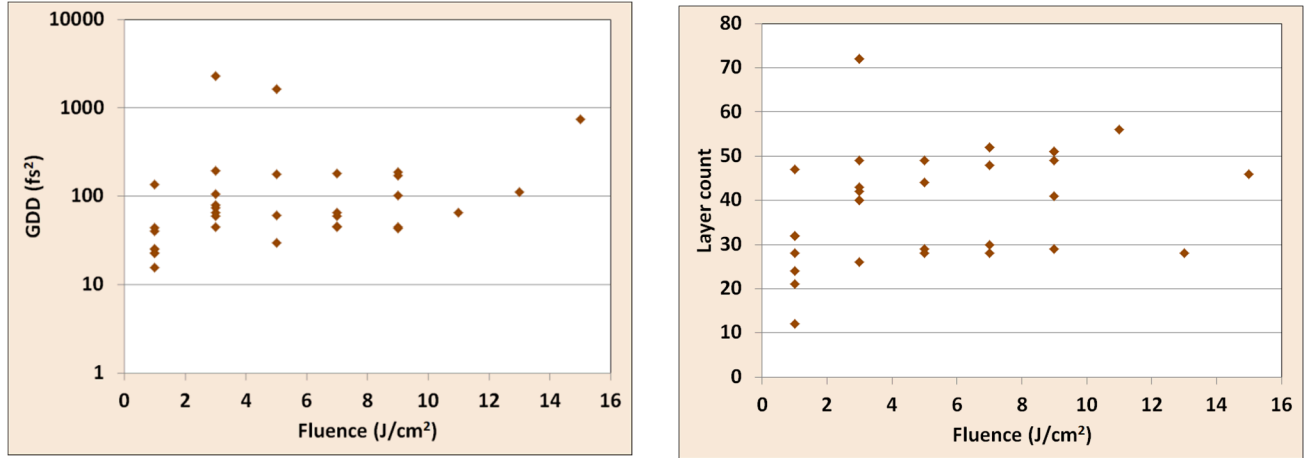


Fig. 6 Short pulse broadband mirror laser resistance as a function of GDD (left image) and layer count (right image).

Multiple coated samples deposited by the same vendor allow for a direct comparison of process differences with a minimal amount of process variation. The laser resistance results of these sister samples, the vendor codes, and the isolated process variables are illustrated in figure 7. When comparing two material coatings deposited at the same vendor, however, with different high index materials, typically the laser resistance decreased as the refractive index increased. Participant J compared e-beam and IAD with the mirror deposited by e-beam showing a significantly higher

laser resistance. One participant submitted coatings using planarization technology to minimize the impact of coating defects.<sup>10</sup> For this pulse length and wavelength, planarized coatings were more laser resistant. Finally there were a few participants that stated that the sister samples were coated under identical conditions. Participant F mentioned that samples 1 & 2 were coated in the same coating run while samples 3 and 4 were also coated together in the same coating run. The consistency of laser damage resistance between sister samples is particularly encouraging. It is unknown to the author whether the other participants who stated that their sister samples were coated in the same coating run or coated in two sequential runs, but under identical conditions. Within this population of samples (E, P, and Q) there is some variability in the laser resistance of sister samples.

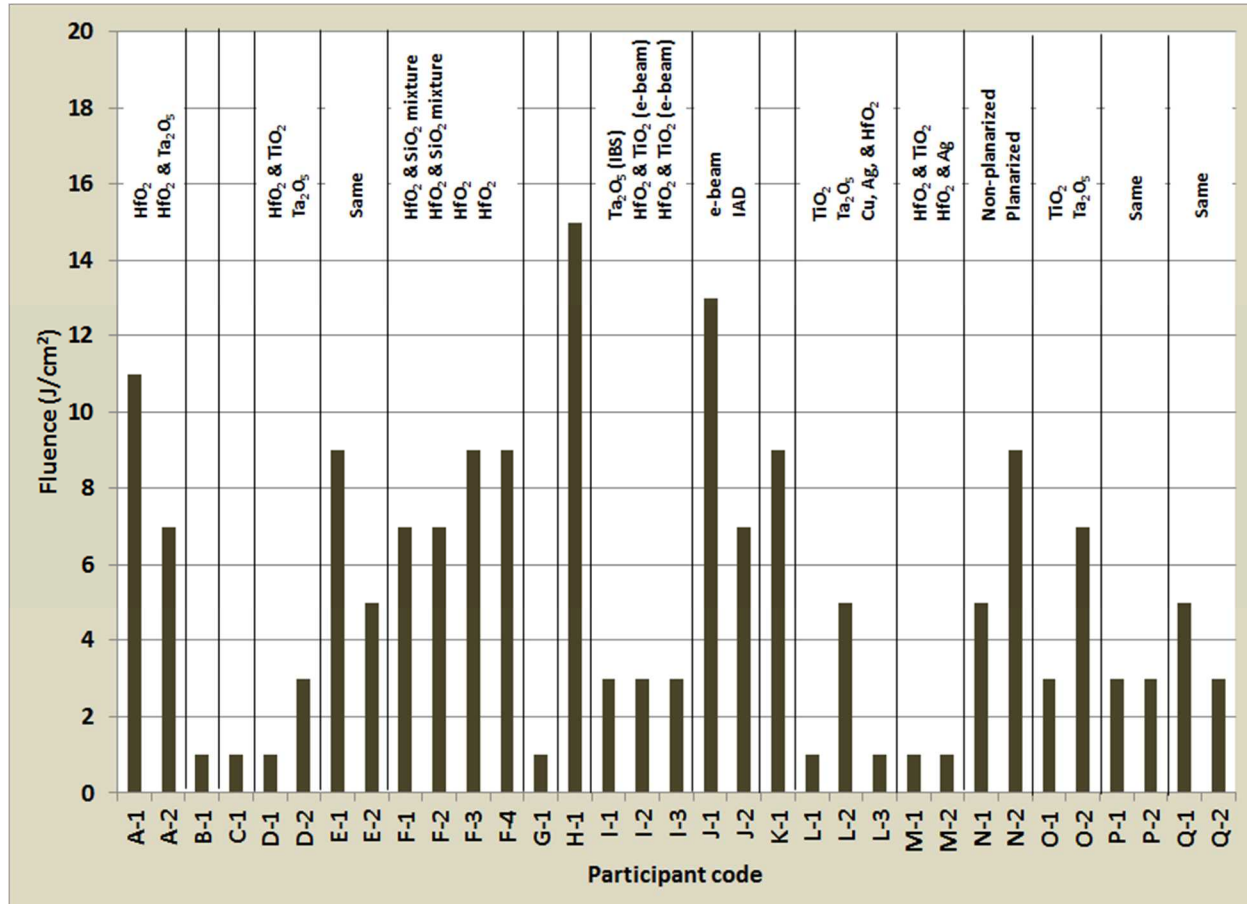


Fig. 7 Laser resistance of the thirty-three samples sorted by participant code.

In order to better understand the impact of high index coating materials and deposition process on the laser resistance of these mirrors, the entire population of samples and the reduced population of samples that meet the GDD specifications are plotted in figure 8. A few global trends can be observed. There is a  $15\times$  fluence difference between the highest and lowest laser resistant samples. In general, two material mirror coatings tended to be more laser resistant than coatings composed of three or more materials. However, the three highest laser resistant samples, which were all two material designs, all failed the GDD specification. These three coatings were all deposited by e-beam, with one sample also having ion assist. Even with optimum centering, the two highest laser resistant coatings had inadequate bandwidth to meet the GDD requirements. The  $11 \text{ J/cm}^2$  e-beam hafnia silica sample actually had sufficient bandwidth, but the GDD was centered at  $54 \text{ fs}^2$  so did not fall within the  $0 \pm 100 \text{ fs}^2$  specification. With elimination of these coatings, the sister samples coated in the same deposition run using ion beam sputtering of hafnia and silica were the highest laser resistant coatings. In a close second came the hafnia silica blend layer design, also coated from the same participant, tied with the IAD zirconia and silica sample and the e-beam three material design consisting of tantala, hafnia, and silica.



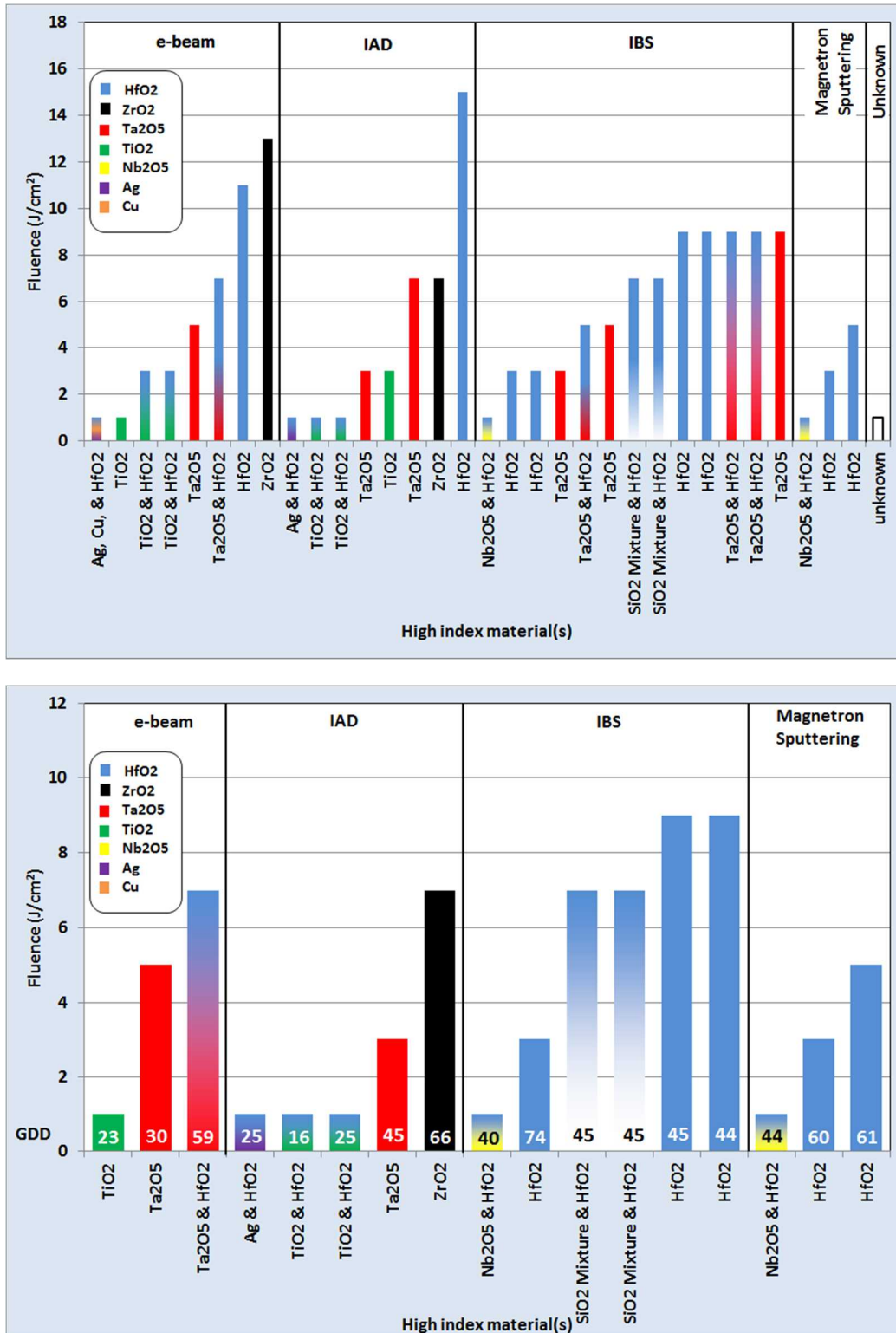


Fig. 8 Laser resistance of the broadband short pulse mirror coatings as a function of coating material, deposition process and GDD for the entire population of samples (top image) and the samples that fully complied with the GDD specifications and reporting criteria (bottom image).

Two of the coated samples contained metallic layers in order to reduce the GDD had very low laser resistance of only 1 J/cm<sup>2</sup>. The silver and hafnia sample had a low GDD of 25 fs<sup>2</sup> which would have been reduced to 16 fs<sup>2</sup> if centered for the minimum GDD. Ironically, the copper, silver, and hafnia sample had a GDD of 136 fs<sup>2</sup> and would have only been reduced to 84 fs<sup>2</sup> if centered for a minimum GDD.

## 6. CONCLUSIONS

As in previous similar thin film laser damage competitions, there was at least an order of magnitude difference between the most and least laser resistant samples and the winner consisted of hafnia and silica, the material combination of choice for high fluence near infrared multilayer coatings. The winning samples were also deposited by ion beam deposition. An unexpected result of this competition is the significant number of samples that were found not to comply with the challenging GDD specifications over a fairly wide spectral range. No correlation was observed between either GDD or layer count and laser resistance.

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## REFERENCES

1. Danson, C., Hillier, D., Hopps N., and Neely D., Petawatt class lasers worldwide, *High Power Laser Science and Engineering*, **3**, e3 doi:10.1017/hpl.2014.52 (2015).
2. Perry, M.D., Pennington, D., Stuart, B.C., Tietbohl, G., Britten, J.A., Brown, C., Herman, S., Golick, B., Kartz, M., Miller, J., Powell, H.T., Vergino, M., and Yanovsky, V., "Petawatt laser pulses", *Optics Letters* **24**, 160-162 (1999).
3. Stolz, C.J., Thomas, M.D., and Griffin, A.J., "BDS thin film damage competition" in *Laser-Induced Damage in Optical Materials: 2008*, G.J. Exarhos, D. Ristau, M.J. Soileau, C.J. Stolz, eds., Proc. SPIE 7132, 71320C-1 - 6 (2009).
4. Stolz, C.J., Ristau, D., Turowski, M., and Blaschke, H., "Thin film femtosecond laser damage competition" in *Laser-Induced Damage in Optical Materials: 2009*, G.J. Exarhos, V. Gruzdev, D. Ristau, M.J. Soileau, C.J. Stolz, eds., Proc. SPIE 7504, 75040S-1 - 6 (2010).
5. Stolz, C. J. and Runkel, J., "Brewster angle polarizing beamsplitter laser damage competition: "S" polarization," in *Laser-Induced Damage in Optical Materials: 2013*, in *Laser-Induced Damage in Optical Materials: 2013*, G.J. Exarhos, V.E. Gruzdev, J.A. Menapace, D. Ristau, and M.J. Soileau, eds., Proc. SPIE 8885, 888509-1-8 (2013).
6. Stolz, C.J., Caputo, M., Griffin, A.J., and Thomas, M.D., "1064-nm Fabry-Perot Transmission Filter Laser Damage Competition" in *Laser-Induced Damage in Optical Materials: 2014*, G.J. Exarhos, V.E. Gruzdev, J.A. Menapace, D. Ristau, and M.J. Soileau, eds., Proc. SPIE 923792370N-1 - 6 (2014).
7. Macleod, H. A., Essential Macleod Thin Film Design Software, <http://www.thinfilmcenter.com/essential.html>
8. Kafka, K.R.P., Chowdhury, E., Negres, R.A., Stolz, C.J., Bude, J.D., Bayramian, A.J., Marshall, C.D., Spinka, T.M., and Haefner, C.L., "Test station development for laser-induced optical damage performance of broadband multilayer dielectric coatings," in *Laser-Induced Damage in Optical Materials: 2015*, G.J. Exarhos, V.E. Gruzdev, J.A. Menapace, D. Ristau, and M.J. Soileau, eds., Proc. SPIE 9632, to be published.
9. Borden, M. R., Folta, J. A., Stolz, C. J., Taylor, J. R., Wolfe, J. E., Griffin, A. J., and Thomas, M. D., "Improved method for laser damage testing coated optics," in *Laser-Induced Damage in Optical Materials: 2005*, Proc. SPIE 5991, 59912A-1-8 (2006).
10. C. J. Stolz, J. E. P. B. Mirkarimi, J. A. Folta, J. Adams, M. G. Menor, N. E. Teslich, R. Soufli, Carmen S. Menoni, and Dinesh Patel, "Substrate and coating defect planarization strategies for high-laser fluence multilayer mirrors," *Thin Solid Films* (2015), to be published.